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<b>(54) Title:</b> BIAXIALLY ORIENTED POLYPROPYLENE BASED MATTE-TRANSLUCENT MULTILAYER FILMS, PROCESS FOR THEIR PRODUCTION AND THE USE THEREOF  <b>(57) Abstract</b>  <p>         Biaxially oriented multilayer matte-translucent film useful for photographic paper laminations, reproduction pictures and menu coatings, conventional lamination purposes, hygienic applications, health care products, food stuffs packaging, cosmetics and picture frames, high speed packaging machines, and as matte tape base films, and for general packaging and overwrapping applications, comprising 15-50 <math>\mu\text{m}</math> layers from (A) 1.5-3.0 <math>\mu\text{m}</math> skin matte-translucent layer containing ethylene(E)-propylene(P)-butylene(B) terpolymer with B-unit content of 1.5-4.9 % and B-unit content of 5.5-15 % polypropylene, high density polyethylene (HDPE) and/or 10:(0.5-1.5) mixture or blend of HDPE with polymethyl(meth)acrylates, alternating or random copolymer of vinyl monomer (styrene, alkyl(meth)acrylates, etc.) or <math>\alpha</math>-olefins (<math>\text{C}_{2-10}</math>) with anhydrides or esters of unsaturated mono- or dicarbonic acids as matting agent, synthetic silica as an antiblocking agent, erucicamide as a slip agent and allyl phosphite (APH) as antioxidant, calcium stearate as a neutralizer, and tetrakis [methylene(3,5-di-<i>tert</i>-butyl-4-hydroxy-hydro-cinnamate) methane as a stabilizer, (B) <math>\geq 0.5 \mu\text{m}</math> polypropylene inner layer containing said APH antioxidant, stabilizer, neutralizer and/or slip agent, and/or <math>\geq 1.5 \mu\text{m}</math> inner layer with composition as a (A) matte layer (C) 10-46 <math>\mu\text{m}</math> virgin or marked polypropylene core layer containing 5-cholesten-3<math>\beta</math>-ol as a marked agent, said APH antioxidant, stabilizer and neutralizer, and/or said slip agent and ethoxylated amine as an antistatic agent, (D) <math>\geq 1.5 \mu\text{m}</math> polypropylene inner layer containing said APH antioxidant, stabilizer and neutralizer, and (E) <math>\geq 0.5 \mu\text{m}</math> outer shining layer containing polypropylene, said antiblocking agent, APH antioxidant and neutralizer, and/or <math>\geq 1.5 \mu\text{m}</math> outer matte layer with composition as a (A) layer, were prepared by using the tandem extruder system supplied with three or four satellite co-extruders, flat die, chill roll, and recycling line. After biaxially stretching and air corona discharged in the given conditions matte-translucent film has sheen (angle 85°) of 10-15 (skin matte side) and 42-61 (outer shining side), longitudinal shrinkage of <math>\geq 0.5</math> % at 90 °C, and <math>\geq 2.5</math> % at 120 °C and transverse shrinkage of <math>\geq 0.25</math> % at 90 °C, and <math>\geq 1.5</math> % at 120 °C for a period at 15 min, excellent matte appearance, surface tension of 40-41 mN/m (after storage for 6 months), high dimensional stability, and low heat sealing temperature of <math>\leq 110</math> °C.       </p>		

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**BIAXIALLY ORIENTED POLYPROPYLENE BASED MATTE-  
TRANSLUCENT MULTILAYER FILMS, PROCESS FOR THEIR  
5 PRODUCTION AND THE USE THEREOF**

The present invention concerns the field of plastics fabrication and uses, specifically, biaxially oriented polypropylene multilayer matte-translucent films.

The packaging food and non-food industry, agriculture, industrial markets, art,  
10 etc. has a high demand for biaxially oriented thin films of different types such as matte, colorless, antifog, antibacterial, photosensitivity, biodegradable, heat-sealable, non heat-sealable, mono- and multilayer, laminated, metallized and other films and materials.

The status of the development of the packaging technology using polyolefins,  
15 further prospects for using polyolefins in the composition of packaging films, advantageous properties and economic aspects of application of biaxially oriented polypropylene films were the object of discussion of several articles and International Conference on polypropylene [Clancioto R., *Packaging* (USA), 1989-suppl.:encycl.-C, 29-31; Jhaveri R., *CEW: Chem. Eng. World*, 24(9), 37(1989); Isaka T., *Kobunshi*, 41(6), 402 (1992); *Chem. Abstr.*, 118, 214372b  
20 (1993); Int. Conf. on Polypropylene-the way ahead, Madrid, 9-10 Nov., Prepr.-London, 1989].

Matte appearance films can be produced by using conventional film-forming polymers such as polyesters, polyamides, polyolefins, vinyl chloride polymers,  
25 styrene polymers, etc. and different methods for preparation of matte surfaces.

Matte surface film for artist's overlays comprises a translucent polymer base film coated with a dried film of matting lacquer comprising: (1) a film-forming plastic resin on the base of styrene-maleate resin and ethyl cellulose, (2) a polystyrene, polyester and polyvinylchloride base film, (3) a relatively soft matting  
30 agent - non elastomeric polyethylene particles, (4) relatively hard matting agent

such as silica and calcium carbonate, and (5) a mixture of organic solvents [Pat. 4719141, USA (1988)].

Several early patents comprise matted biaxially oriented polyethylene terephthalate film preparing by using water, matting solution (10-20 %) of  
5 diethylene glycol-ethylene glycolterephthalic acid copolymer in formal glycol, mixture of melamine resin and silicic acid, 2,4-dichlor-6-hydroxy-s-triazine (0.05 %) in water, and also polystyrene [Pat. 434094, SU (1974), Pat. 265855, SU (1976), Pat. 301841, SU (1976), Pat. 49-132166, Japan (1974)]. For example, polyethylene terephthalate (PET) film with a matte surface for drawing office  
10 applications containing polystyrene (1-20 %) and  $\text{TiO}_2$  (1-10%) as a filler was biaxially oriented at 105-110 degree by stretching. The material prepared has 60-65 % transmission and a pencil line on it has density of  $0.7\text{-}0.8 \text{ g/cm}^3$  [Pat. 434094, SU (1974)].

Matte surface polymer film useful for preparation of drawing papers, magnetic  
15 recording tape leader, wallpaper, capacitor insulators, labels, etc., were prepared from polyester-PET (80 parts), polystyrene (10 parts) and polymethylmethacrylate (10 parts) by stretching 150 % mono- or biaxially and optionally, laminated on one side of the polyester films. The film obtained has good mechanical strength and writing ability [Pat. 63-193920, Japan (1974)].

20 Matte polyamide film contains itaconic acid-methyl acrylate-vinylidene chloride terpolymer (0.1-10 parts) as a matting agent and have good antiblocking properties without sacrificing heat sealability [ *Chem. Abstr.*, 80 (12), 60651z].

Biaxially stretched anisotropic films comprising ethylene-propylene copolymer or blends of polypropylene and polyethylene (1-10 %) were prepared by extruding  
25 at 260°C and sprayed onto a casting roll at 30°C, stretched longitudinally 5.6 times at 145°C and transversely 9.3 times at 157°C [Pat. 1936963, Germany (1971)].

New film developments by using modern manufacturing processes such as stenter or flat die and bubble or tubular processes and productivity improvements were allowing biaxially oriented polypropylene films to pierce through new  
30 markets as replacement for conventional flexible packaging materials such as

paper, aluminium foil, and other plastic films (PET, PA, PVC, LDPE, HDPE, cellulose, etc.).

Several patent publications relate to polyolefin (homo- and copolymers of propylene with  $\alpha$ -olefins) based multilayer transfer metallization films, including  
5 also matte and/or opaque appearance films [Pat. 63-73146 (Japan), Pat. EP-A-O 175259 and EP-A-O 352463, Pat. Appl. 95 / 936 (Turkey), Pat. 4135096 (Germany), Pat. 88048244 (UK) and 88199666 (UK)]. Thus, Pat. 4135096 (Germany) comprises a multilayer matte appearance film containing a polypropylene based layer and an outlayer made from an HDPE and propylene  
10 homo-, copolymer and terpolymer blends which has minimum sheen and maximum haze.

Mitsubishi Royan Co., Ltd. the process for producing of translucent polypropylene monolayer film was patented [Pat. 41-38520, Japan (1979)]. Other recently publication Japan patent relates to a matte packaging film comprising  
15 polypropylene and Al borate as translucency-providing agent which was prepared from mixture of 90 parts of polypropylene and 10 parts of Alborex Y (AL borate whisker) by extruding through a temperature-die. The translucent film with 30  $\mu$ m thickness have an adequate surface unevenness [Pat. 06-212037, Japan (1994)].

The object of Pat. EP-A-O 367613 was a multilayer film containing a vacuole  
20 polypropylene base layer and an incribable outer layer made from a mixture or blend of HDPE with polyolefins. The title film has a density of  $0.69 \text{ g/cm}^3$ , opaque appearance and non-transparent to light properties. Additionally, it was preferred to incorporate a  $\text{SiO}_2$ -filler into the outer layer to improve the incribability. This layer was also very matte, which gives the film a paper-like appearance.  
25 Therefore,  $\text{SiO}_2$  containing matte surface proves to be unsatisfactory on the printing.

Biaxially oriented multilayer ( $\leq 3$  layers) polyolefin films including polypropylene and HDPE, and having low coefficient friction, excellent optical transparency, good wettability, high sheen and low haze were invented in the  
30 patents of 4419411 (USA), 4578316 (USA) and 89/10839 (WO).

The object of Pat. 5478643, USA (1995) was a 21.5  $\mu\text{m}$  thickness matte multilayer ( $\leq 3$  layers) polypropylene film for use in a metallization process which contains one base polypropylene layer (19  $\mu\text{m}$  thickness) with migrating additives such as N,N-bis-hydroxyethyl) ( $\text{C}_{10-20}$ )alkylamine (0.22 %) and stearamide (0.25 %)  
5 % and each outer layer (1.25  $\mu\text{m}$  thickness) including a mixture or blend of polypropylene, ethylene-propylene copolymer and/or ethylene-propylene-butylene terpolymer, and HDPE, blend of HDPE with said propylene copolymer or terpolymer. This outer layer was distinguished by a characteristic matte surface or appearance and was suitable for use as a transfer metallization film. All layers of  
10 said film contained 0.12 % by weight of pentacrythrityl tetrakis[4-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate] as a stabilizer and 0.05 % by weight of calcium stearate as a neutralizer. The film resulted was then co-extruded the melts corresponding to the individual layers through a flat-film die, taken the film off over a take-off roll at 40-100°C, biaxially stretched at a longitudinal stretching  
15 ratio about 4:1 to 7:1 and a transverse stretching ratio about 8:1 to 10:1, heat setting and corona treated (one or both outer layer). The film prepared has following average characteristics: thickness 10-50  $\mu\text{m}$ , density 0.90-0.92  $\text{g}/\text{cm}^3$ , sheen (85° angle) 29-42, haze 52-70 %, roughness 2.3-3.2  $\mu\text{m}$ , coefficient of friction 0.3-0.5, surface tension (after storage for 6 months) 38-39  $\text{mN}/\text{m}$ ,  
20 and very good matte appearance (visual assessment). Similarity of said patent with the present invention that the film consists HDPE, polypropylene and ethylene-propylene-butylene terpolymer; the film in the both cases have a matte appearance. Difference and deficiency of said film are the following: the film mainly were recommended for use in metallization process; co-extruder system  
25 used allows only to prepare maximum 3-layer film; the film not contains a matting agent such as a carbonyl containing alternating or random copolymer, and also have comparative high values of sheen, and low values of surface tension (Table 1).

Pat. 5489454, USA (1996) describes a biaxially oriented, shrinkable, heat-sealing 3-layer (10-50  $\mu\text{m}$  thickness) polyolefin film with a matte appearance,  
30

good passage through the machine and antistatic and shrink properties, which simultaneously forms very tight, effective bil-low- and fold-free wrapping, and also was provided a co-extruding process for its production. In order to improve the specific properties of these polyolefin films both the base layers  
5 (polypropylene or polypropylene mixture) and outer layer(s) (mixture or blend of propylene copolymer or terpolymer and HDPE) contain 0.1-0.2 %  $\text{SiO}_2$ ,  $\text{CaCO}_3$  and/or incompatible organic polymers such as polyamides, polyesters, polycarbonates, benzoguanamine-formaldehyde resins as antiblocking agents, 0.05-2.0 % phenolic stabilizers, 0.05-0.25 % high aliphatic acid amide and  
10 polydimethyldisiloxanees as lubricants, 0.05-0.3 % glycerol monostearate as antistatic agent. The film resulted was stretched in the longitudinal direction preferably from about 4:1 to 9:1 at 80-120°C and in the transverse direction from about 6:1 to 8:1 at 130-155°C. The films with different compositions has the following characteristics: sheen (85° angle) 30-38, haze 62-68 %, roughness 2.6-  
15 3.0  $\mu\text{m}$ , coefficient of friction 0.3-0.4, surface tension 39  $\text{mN/m}$ , minimum heat-sealing temperature 105-115°C, longitudinal and transverse shrinkage at 90°C 4-6 % and 3-5 % at 120°C 11-14 % and 16-20 % respectively. Similarity is in the fact that the use in film composition of propylene homo- and terpolymer, and HDPE; difference and deficiency are the following: the films contained  $\text{SiO}_2$ ,  
20  $\text{CaCO}_3$  and/or said incompatible organic polymers as matting and antiblocking agents and have comparative high values of sheen, and longitudinal and transverse shrinkage and low values of surface tension (Table 1).

Three early patents (DE-A 1694694, US 4419411 and EP-A 008904) were related to biaxially oriented polyolefin multilayer films having good heat-sealing  
25 on the both sides. However, these films has low surface and mechanical properties, and also unsatisfactory processing properties in high-speed packaging machines.

Recently published Pat. 5492757, USA (1996) discloses a opaque-matte four-layer film (28  $\mu\text{m}$  thickness) of A/B/C/D structure comprising (B) base layer  
30 (22.7  $\mu\text{m}$  thickness) from a polypropylene, 4.2-5.0 %  $\text{CaCO}_3$  filler and 3.2-8.0 %

TiO<sub>2</sub> filler, (C) interlayer (3.5 µm thickness) from a 1:1 mixture of ethylene(1.9 %)-propylene-butylene(8.4 %) terpolymer and HDPE, (D) outer layer (0.9 µm thickness) from a said terpolymer with analogous composition and (A) outer layer (0.9 µm thickness) a ethylene-propylene copolymer. All layers contained  
5 conventional stabilizer and neutralizer. The base layer furthermore contained aliphatic alkylamine as antistatic agent. After co-extrusion, extruded four-layer film was taken off a first take-off roll and further trio of rolls. The film was then cooled, subsequently stretched longitudinally and transversely, set and corona treated. The films resulted has matte appearance with gloss (85° angle, D side) 29-  
10 84, coefficient of friction 0.4-0.6, whiteness (D side) 65-81, opacity (D side) 67-70 and density 0.69-0.94 %. The multilayer film according to this patent invention was distinguished by a multiplicity of advantageous properties and was recommended for wide variety of applications such as an attractive matte lamination film, an opaque packaging film in high-speed packaging machines, an  
15 opaque film in wrapping film. and a base film for adhesive tape or for aqueous barrier coating systems. The film was also suitable for the production of plastic labels and laminates with paper, cardboard, metals, and metalized plastic films. The comprise of properties these films with properties of films according to present invention (Table 1) shown that the films of said invention have high value  
20 of gloss of matte side. The film contain only HDPE as matting agent and terpolymer with E- and B-units differing from composition of terpolymer using in present invention.

High shrinkage polyolefin multilayer films were disclosed in the following early patent publications: EP-A-O 436196, EP-A-O 350859, DE-A 1954467 and DE-A  
25 3215120. The films according to these patents has very high sheen values and very low haze. Therefore these shrink films were unsatisfactory with respect to their passage through the machines, and their properties during winding, further processing and slitting.

Pat. 5494717, USA (1996) relates to a biaxially oriented matte polypropylene  
30 film of high shrinkage which comprises a three-layers of X/Z/X structure, i.e., one



base layer Z containing a polypropylene or propylene polymer mixture and two outer layers X containing mixture or blend of polypropylene, ethylene-propylene copolymer ethylene-propylene-butylene terpolymer and HDPE. In order to improve specific properties of films both the base and outer(s) layers include  
5 conventional additives such as known antiblocking and antistatic agents, stabilizers, neutralizers and lubricants. High shrinkage, uniform matte appearance, good passage through the machine, good antistatic properties and high suitability for rolling up of films resulted allow to use as film packaging for books, frames, toys, food stuffs, cosmetics, etc., and also for metallization and lamination after  
10 corona or flame treatment. The films prepared after biaxially oriented and corona treated has sheen 29-40 (85° angle), has 58-70 %, roughness 2.3-3.2  $\mu\text{m}$ , coefficient of friction 0.3-0.5, surface tension 38-39  $\text{mN/m}$ , longitudinal and transverse shrinkage at 120°C 29-36 % and 41-47 % respectively.

Analogously with present invention homo- and terpolymer of propylene, HDPE,  
15 antiblocking agent and neutralizer are used in the composition of corona-treated biaxially oriented films. The films of present invention as compared with matte film of said patent have advantageous properties (Table 1).

The object of several patent publications [EP-A-O 479101, EP-A-O 432452 and DE-A 3535472] was multilayer transparent twist wrapping films made from  
20 mono- and/or biaxially balanced oriented propylene polymer and copolymers. Thus, Pat. EP-A-O 479101 (1992) discloses a biaxially oriented polypropylene film (25  $\mu\text{m}$  thickness) for twist-wrapping which was prepared from mixture of isotactic polypropylene (71.6 %), hydrogenated cyclopentadiene resin with softening point 140°C (28 %), ethoxylated amine (0.2 %) as an antistatic agent,  
25 erucicamide (0.2 %) as a slip agent, and polydimethylsiloxane (0.5 %) as a surfactant by co-extruding and drawn longitudinally 6.5-fold at 110°C and transversely 7.3-fold at 150°C. After corona treatment said film had surface tension 40  $\text{mN/m}$ , and elastic modulus 3200 and 3400  $\text{N/mm}^2$ , tensile strength 20.9 and 21.9  $\text{kg/mm}^2$ , elongation 95 and 78 %, shrinkage 11 and 7 % in the  
30 longitudinal and transverse directions, respectively. Similarity is in the fact that

corona treated biaxially oriented polypropylene film contains antiblocking agent; difference and deficiency of this film as compared with films of present invention are following: non-filled transparent films contain polydimethylsiloxane as surfactant, and hydrogenated cyclopentadiene resin and have relatively low elongation and high values of shrinkage. These twist wrapping films has unsatisfactory appearance, processing properties and in particular scratch resistance. The aim of Pat. 5496600, USA (1996) was the attempt to remove these deficiencies. Said patent describes a three-layer biaxially oriented polypropylene matte film having advantageous properties such as a characteristic matte sheen at less than 80, very good printability and twist properties, uniform matte appearance and excellent scratch resistance and using as a twist wrapping film. The title film contains homo-, co- and terpolymer of propylene, and HDPE or blend of HDPE with said polyolefins, and also petroleum, terpen and other hydrocarbon resins, including dicyclopentadiene polymer, as amorphous matting agents. The film also has a permanent set of greater than about 50 %, in the longitudinal and transverse directions and approximately isotropic mechanical properties. Heat-setting, biaxially stretched and corona treated film has overall thickness 10-80  $\mu\text{m}$ , density 0.90-0.95  $\text{g/cm}^3$ , sheen 29-42, has 58-70 %, roughness 2.3-3.2  $\mu\text{m}$ , coefficient of friction 0.3-0.5, surface tension 38-39  $\text{mN/m}$ , permanent set 62-64 % and 60-63 % in the longitudinal and transverse directions respectively, and scratch resistance ( $\Delta\text{haze}$ ) 20-22. However, these films as compared with films of present invention have high sheen of matte side, and high values of longitudinal and transverse shrinkage at 120°C (Table 1).

The films of present invention as compared with commercial matte film also have advantageous properties (Table 1).

Thus, it is an object of the present invention to design and prepare a multilayer structure for biaxially oriented polypropylene based matte-translucent films with high sheen, excellent matte appearance surface, lower longitudinal and transverse shrinkage for dimensional stability, low heat sealing temperature, low water

absorption, good lay-flat characteristics, for dimensional stability, resistance to oil, crease and chemicals.

One object of the present invention is to improve the production process of said films by using tandem extruder system supplied with three co-extruders,  
5 recycling line for the film forming in the transverse stretching stage and corona discharge, and also modified variant of chill-roll treatments of films in the biaxially orientation process allowing to form homogen matte films with improved surface properties and dimensional stability.

Another aspect of the present invention to use new polymer system, i.e. mixture  
10 or blend of HDPE with carbonyl containing homo- and copolymers such as polymethyl(metha)acrylates, alternating or random copolymers of vinyl monomers or  $\alpha$ -olefins ( $C_{2-10}$ ) with anhydrides or esters of unsaturated mono- or dicarbonic acids as mattering agent in the skin matte layer of said films.

It is further object of the present invention to widen the field of application  
15 films prepared useful for photographic paper laminations, reproduction pictures and menu coatings, conventional lamination purposes, hygienic applications, health care products, food stuffs packaging, cosmetics and frames, high speed packaging machines, and as matte tape base films, and for general packaging and overwrapping applications.

20 An offered A/B/C/D/C multilayer film of the present invention is biaxially oriented matte-translucent polypropylene based film structure comprising 15-50  $\mu\text{m} \leq 5$  layers from (A) Pat.  $\leq 3.0 \mu\text{m}$  skin matte-translucent layer containing ethylene(E)-propylene(P)-butylene(B) terpolymer with E-unit content of 1.5-4.9 % and B-unit content of 5.5-15 %, polypropylene, high density polyethylene  
25 (HDPE) and/or 10: (0.5-1.5) mixture or blend of HDPE with polymethyl(metha)acrylates (mol. weight  $M_n = (55-160) \times 10^3$ ), alternating or random copolymer of vinyl monomer (styrene, alkylacrylates or methacrylates, etc.) or  $\alpha$ -olefins ( $C_{2-10}$ ) with anhydrides or esters of unsaturated mono- or dicarbonic acids (mol. weight  $M_n = (10-200) \times 10^3$ ) as mattering agent, synthetic  
30 silica as an antiblocking agent, erucicamide as a slip agent and allyl phosphite

(APH) as antioxidant, calcium stearate as a neutralizer, and tetrakis[methylene (3,5-di-*tert*-butyl-4-hydroxy-hydro-cinnamate)] methane as a stabilizer, (B)  $\geq 0.5$   $\mu\text{m}$  polypropylene inner layer containing said APH antioxidant, stabilizer, neutralizer and/or slip agent, and/or  $\geq 1.5$   $\mu\text{m}$  inner layer with composition as in  
5 (A) matte layer, (C) 10-46  $\mu\text{m}$  virgin or marked polypropylene core layer containing 5-cholesten-3 $\beta$ -ol as a marked agent, said APH antioxidant, stabilizer and neutralizer, and/or said slip agent and ethoxylated amine as a antistatic agent, (D)  $\geq 1.5$   $\mu\text{m}$  polypropylene inner layer containing said APH antioxidant, stabilizer and neutralizer, and (E)  $\geq 0.5$   $\mu\text{m}$  outer shining layer containing  
10 polypropylene, said antiblocking agent, APH antioxidant and neutralizer, and/or  $\geq 1.5$   $\mu\text{m}$  outer matte layer with composition as a (A) layer.

According to the present invention the technological aspect of manufactured process of said films is distinguished from known processing used in the production of matte films by the fact that tandem extruder system with two main  
15 extruders supplied with three satellite co-extruders, recycling line and corona discharge. The process is carried out by three chill-roll treatments and two step of longitudinal orientation allowing to prepare good homogenized matte film with improving surface properties and dimensional stability. One or both surface of biaxially oriented films prepared are treated in a known manner by air corona  
20 discharge. The use of said recycling line for film waste forming in the transverse stretching stage allows to lower film cost by 3.5 %.

After coextrusion, the extruded five-layer film is taken off over the corresponding process steps through a chill roll and cooled, and cast film profile is controlled by  $\beta$ -Gauge equipment. The film is subsequently stretched  
25 longitudinally at two steps and stretched transversely. After biaxially orientation, the film is set and air corona-treated on one or two sides. The following conditions in detail, are selected: (1) Extrusion: extrusion temperature 200-260°C, first chill roll temperature 35-45°C; (2) Longitudinal stretching: stretching roll temperature of first step 150-160°C and second step 140-150°C, longitudinal stretching ratio  
30 5:1 - 6:1 for first step and 1:1 - 1:2 for second step; Transverse stretching:

temperature of heat-up zones 170-185°C, temperature of stretching zones 160-170°C, transverse stretching ratio 9:1 - 10:1; Recycling: edges of the biaxially orientated film is recycled and fed to the line again; Setting: setting temperature 165-170 °C; Air corona discharge: voltage 15-25 kV and frequency 20-30 kHz.

- 5 The following Examples of the present invention for preparation of multilayer matte-translucent films with different composition, properties are illustrated.

### Example 1

- A/B/C/D/E structure film with a thickness of 17.5 µm comprising (A) 3.0 µm  
 10 skin matte-translucent layer from 47.9 % by weight of ethylene(E)-propylene(P)-butylene(B) terpolymer with E-unit content of 3.5 % and B-unit content of 5.5 % (density 0.90 g/cm<sup>3</sup>, MFI 8.0 g/10 min, m.p. 143.6°C, vicat softening point 120°C, tensile strength 23 MPa, elongation at yield 13 %), 49.9 % by weight of high density polyethylene (HDPE, density 0.964 g/cm<sup>3</sup>, MFI 3.5 g/10 min, m.p.  
 15 134°C, vicat softening point 128°C, Izod impact strength 35 kg-cm/cm, tensile strength 28.4 MPa, elongation at break 500 %) as mattering agent, 1.8 % by weight of polypropylene (density 0.905 g/cm<sup>3</sup>, MFI 3.5 g/10 min at 230°C and load at 21.6 N, m.p. 163°C, vicat softening point 150°C, Izod impact strength 44 kg-cm/cm, tensile strength 32 MPa, elongation at break 700 %), 0.25% by weight of  
 20 synthetic silica as an antiblocking agent, 0.05 % by weight of erucicamide as a slip agent, 0.05 % by weight of allyl phosphite (APH) as antioxidant, 0.1 % by weight of calcium stearate as a neutralizer, (B) 0.5 µm inner layer from 99.87 % by weight of polypropylene, 0.04 % by weight of allyl phosphite (APH) as a antioxidant, 0.04 % by weight of tetrakis[methylene (3,5-di-*tert*-butyl-4-hydroxy-  
 25 hydro-cinnamate] methane as a stabilizer, 0.05 % by weight of calcium stearate as a neutralizer, (C) 13 µm core layer from virgin or marked polypropylene (0.01% of 5-cholesten-3β-ol as a marked agent) with composition as in (B) layer, (D) 0.5 µm polypropylene inner layer with composition as in (B) layer and (E) 0.5 µm outer shining layer from 99,7 % by weight of polypropylene, 0.2 % by weight of  
 30 said antiblocking agent, 0.05 % by weight of APH antioxidant and 0.05 % by

weight of said neutralizer, was prepared by using the tandem extruder system supplied with four satellite co-extruders, flat die, chill roll, recycling line. After biaxially stretching, setting and air corona discharged in the given conditions the film prepared has composition and properties showing in Tables 2, 3 and 4  
5 respectively.

### Example 2

The film with a thickness of 50  $\mu\text{m}$  and A/B/C/D/E structure as in Example 1 repeated with following changes: (A) matte-translucent skin layer comprises a E-P-B terpolymer with E-unit content of 3.5 % and B-unit content of 5.5 %, thickness of (C) core layer is 45.5  $\mu\text{m}$ ; (E) 0.5  $\mu\text{m}$  outer layer is matte containing a E-P-B terpolymer with E-unit content of 4.2 % and B-unit content of 9.0 % and with composition as in (A) layer. After biaxially stretching, setting and air corona discharged in the given conditions the film prepared has  
15 composition and properties showing in Tables 2, 3 and 4 respectively.

### Example 3

The film with a thickness of 30  $\mu\text{m}$  and A/B/C/D/E structure as in Example 1 repeated with following change: thickness of (C) core layer is 25.5  $\mu\text{m}$  and  
20 contains ethoxylated amine as an antistatic agent. After biaxially stretching, setting and air corona discharged in the given conditions 3.0 $\mu\text{m}$  / 0.5 $\mu\text{m}$  / 25.5 $\mu\text{m}$  / 0.5  $\mu\text{m}$  / 0.5 $\mu\text{m}$  film prepared has composition and properties showing in Tables 2, 3 and 4 respectively.

25

### Example 4

The film with a thickness of 30  $\mu\text{m}$  and A/B/C/D/E structure as in Example 3 repeated with following changes: (E) shining outer layer comprises a E-P-B terpolymer with E-unit content of 1.5 % and B-unit content of 15 % (MFI 8.0 g/10  
30 min, m.p. 115°C), erucicamide as a slip agent. After biaxially stretching, setting

and air corona discharged in the given conditions 3.0 $\mu$ m / 0.5 $\mu$ m / 25.5 $\mu$ m / 0.5 $\mu$ m / 0.5 $\mu$ m film prepared has composition and properties showing in Tables 2, 3 and 4 respectively.

5

#### Example 5

The film with a thickness of 30  $\mu$ m and A/C/D/E structure without (B) inner layer having the composition as in Example 1. After biaxially stretching, setting and air corona discharged in the given conditions 3.0 $\mu$ m / 26 $\mu$ m / 0.5 $\mu$ m / 0.5 $\mu$ m film prepared has composition and properties showing in Tables 2, 3 and 4 respectively.

10

#### Example 6

The film with a thickness of 30  $\mu$ m and A/C/D/E structure without (B) inner layer having the composition as in Example 5 repeated with following changes: (C) core layer contains erucicamide as a slip agent and ethoxylated amine as an antistatic agent. After biaxially stretching, setting and air corona discharged in the given conditions 3 $\mu$ m / 26 $\mu$ m / 0.5 $\mu$ m / 0.5 $\mu$ m film prepared has composition and properties showing in Tables 2, 3 and 4 respectively.

15

20

#### Example 7

The film with a thickness of 30  $\mu$ m and A/C/D/E structure without (B) inner layer having the composition as in Example 5 repeated with following changes: (E) shining outer layer contains a E-P-B terpolymer with E-unit content of 1.5 % and B-unit content of 15 %, erucicamide as a slip agent. After biaxially stretching, setting and air corona discharged in the given conditions 3 $\mu$ m / 26 $\mu$ m / 0.5 $\mu$ m / 0.5 $\mu$ m film prepared has composition and properties showing in Tables 2, 3 and 4 respectively.

25

30

### Example 8

The film with a thickness of 15  $\mu\text{m}$  and A/C/E structure having the composition as in Example 1 without (B) and (D) inner layers. After biaxially stretching, setting and air corona discharged in the given conditions 3.0 $\mu\text{m}$  / 11 $\mu\text{m}$  / 1.0 $\mu\text{m}$  film prepared has composition and properties showing in Tables 2, 3 and 4 respectively.

### Example 9

The film with a thickness of 17.5  $\mu\text{m}$  and A/B/C/D/E structure as in Example 1 repeated with following changes: (A) matte-translucent skin layer comprised a E-P-B terpolymer with E-unit content of 4.2 % and B-unit content of 9.0 %, 10:1 mixture or blend of HDPE with alternating styrene-maleic anhydride copolymer (St-MA, intrinsic viscosity,  $\eta_{\text{in}}$  1.2 dl/g at 20°C in dimethylformamide, molecular weight  $M_n$  2.0  $\times 10^5$ , acid number 550 mg KOH/g, softening point 215°C) as a mattering agent. After biaxially stretching, setting and air corona discharged in the given conditions the film prepared has composition and properties showing in Tables 2, 3 and 4 respectively.

### Example 10

The film with a thickness of 17.5  $\mu\text{m}$  and A/B/C/D/E structure as in Example 1 repeated with following changes: (A) matte-translucent skin layer comprised a E-P-B terpolymer with E-unit content of 4.2 % and B-unit content of 9.0 % containing said slip and antiblocking agents (density 0.885 g/cm<sup>3</sup>, MFI 7.3 g/10 min, m.p. 126 °C, vicat softening point 113°C, Izod impact strength 8.7 kg-cm/cm, tensile strength 20 MPa, elongation at break 560 %), 10:1 mixture or blend of HDPE with polymethylmethacrylate (PMMA, density 1.185 g/cm<sup>3</sup> at 25°C, Tg 376°C by DSC, molecular weight  $M_n$  1.5  $\times 10^5$ ,  $\eta_{\text{in}}$  1.52 dl/g at 20°C in acetone) and/or PMMA/polypropylene mixture as a mattering agent. After biaxially stretching, setting and air corona discharged in the given conditions the film prepared has composition and properties showing in Tables 2, 3 and 4 respectively.



### Example 11

The film with a thickness of 50 $\mu$ m and A/B/C/D/E structure as in Example 9 repeated with following changes: (A) matte-translucent skin layer comprised a E-  
5 P-B terpolymer with E-unit content of 3.5 % and B-unit content of 5.5 % and 10:1  
mixture or blend of HDPE /  $\alpha$ -diisobutylene-maleic anhydride  
alternating copolymer (softening point 195°C, acid number 665 mg KOH/g,  $\eta_{in}$   
0.13 dl/g at 20°C in methylethyl ketone). After biaxially stretching in the given  
conditions, setting and air corona treatment 3.0 $\mu$ m / 1.5 $\mu$ m / 43 $\mu$ m / 1.0 $\mu$ m /  
10 1.5 $\mu$ m film prepared has composition and properties showing in Tables 2, 3 and  
4 respectively.

For analysis of the initial material used and films prepared following known  
standard measurement methods are used:

15

Density is determined in according to ISO 1183 and/or ASTM D-1505.

Melting Flow Index (MFI) is measured in according to ASTM 1238 / L at  
230°C and load of 21.6 N.

20 Melting point (m.p.) is measured by DSC method, maximum point of the  
melting curve, at heating rate 10°C/min is corresponded to m.p. value.

Vicat softening point is determined in according to ASTM D-1525.

25 Izod impact strength is measured in according to ISO 180 /1A.

Tensile strength and Elongation are determined in according to ASTM D-882.

Sheen of the film is measured in accordance with ASTM D-2103, the angle of  
incidence is set at 85°.

30 Haze of the film is measured in accordance with ASTM D-1003.

Coefficient of friction of the film is determined in accordance with ASTM D-  
1984.

Shrinkage of the film is measured in accordance with ASTM D-1204. The test sample is shrink at 90 and 120°C for a period at 15 *min*. Longitudinal shrinkage (LS) and transverse shrinkage (TS) are than given as the difference between the  
5 elongation determined ( $L_t$  and  $T_t$ ) compared with the initial lengths ( $L_o$  and  $T_o$ ) by following equation:  $LS = (L_o - L_t) / L_o$  and  $TS = (T_o - T_t) / T_o$ .

Matte appearance and optical quality of the film are assessed visually ( + good, ++ very good and +++ excellent).

10

Surface tension of the film after surface ionization by air corona discharge is measured in accordance with ASTM D-2578.

Table 1. Comparative Properties of Present Invention, Known Patents  
and Commercial Matte Films

Properties	Present Invention			Known Patents*						Commer- cial Film
	3-layers	4-layers	5-layers	[1]	[2]	[3]	[4]	[5]	[6]	
Thickness ( $\mu\text{m}$ )	15	30	30	25	10-50	10-50	50-100	5-40	10-80	15
Density ( $\text{g}/\text{cm}^3$ )	0.89	0.90	0.90	-	0.92	0.92	$\leq 0.94$	0.94	0.95	0.89
Sheen (angle $85^\circ$ ) Skin matte surface	13	15	19	-	29-42	30-38	29-84	29-40	29-42	27
Haze ( % )	55	62	70	-	60-70	62-68	-	58-70	60-70	56
Coefficient of friction	0.5	0.6	0.3	-	0.4	0.35	0.5	0.4	0.4	0.3
Shrinkage ( % )										
90°C in LS	1.0	0.5	1.25	-	-	4-6	-	-	-	-
90°C in TS	0.25	0.4	0.5	-	-	3-5	-	-	-	-
120°C in LS	3.0	2.5	5.5	-	-	11-14	-	29-36	-	2.75
120°C in TS	1.5	2.0	2.25	-	-	16-20	-	41-47	-	1.0
Tensile strength ( $\text{kg}/\text{mm}^2$ ) in LS	10	12	15	20.9	-	-	-	-	-	10.7
in TS	21	25	30	21.9	-	-	-	-	-	23.6
Elongation ( % )										
in LS	135	150	170	95	-	-	-	-	-	170
in TS	43	52	61	78	-	-	-	-	-	60
Minimum heat sealing temp.(°C)	95	110	105	-	-	105-115	-	-	-	135
Matte appearance (visual assessment)	+++	+++	+++	-	-	--	-	++	++	--
Surface tension** ( $\text{mN} \cdot \text{m}$ )	40	41	41	-	38-39	38-39	-	38-39	38-39	37

\* [1] - Pat. EP-A-O 479101, [2] - Pat. US 5478643, [3] - Pat. US 5489454,  
[4] - Pat. US 5492757, [5] - Pat. US 5494717, and [6] - Pat. US 5496600.

\*\* Measured after storage of corona discharged films for 6 months.

Table 2. Compositions of Biaxially Oriented Polypropylene Based  
Matte-Translucent Films of Present Invention

Components and Additives	Composition of Films (%)										
	E1	E2	E3	E4	E5	E6	E7	E8	E9	E10	E11
Polypropylene	83.06	93.00	89.86	89.60	90.03	89.88	89.64	80.21	83.07	83.07	93.98
E-P-B terpolymer (I)*	8.21	2.87	4.78	-	4.80	4.80	4.80	9.60	-	-	2.90
E-P-B terpolymer (II)*	-	0.48	-	-	-	-	-	-	8.28	8.28	-
E-P-B terpolymer (III)*	-	-	-	5.21	-	-	0.40	-	-	-	-
Polyethylene (HDPE)	8.56	3.49	5.00	5.00	5.00	5.00	5.00	10.00	7.71	7.71	2.70
St-MA copolymer	-	-	-	-	-	-	-	-	0.77	-	-
Polymethylmethacrylate	-	-	-	-	-	-	-	-	-	0.77	-
DIB-MA copolymer	-	-	-	-	-	-	-	-	-	-	0.27
Erucicamide (EA)	0.01	0.005	0.005	0.006	0.005	0.005	0.006	0.01	0.01	0.01	0.003
Synthetic silica (SS)	0.04	0.02	0.02	0.024	0.02	0.02	0.024	0.05	0.04	0.04	0.02
Calcium stearate (Cast)	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.06	0.05	0.05	0.05
Tetrakis methane (TAM)**	0.03	0.04	0.05	0.055	0.035	0.035	0.035	0.03	0.03	0.03	0.036
Allyl phosphite (APH)	0.04	0.045	0.065	0.055	0.055	0.04	0.04	0.04	0.04	0.04	0.04
Ethoxylated amine(EtA)	-	-	0.17	-	-	0.17	-	-	-	-	-

\* Terpolymer with following ethylene(E) and butylene(B) unit contents :

- (I) E-unit content of 3.5 % and B-unit content of 5.5 %
- (II) E-unit content of 4.2 % and B-unit content of 9.0 %
- (III) E-unit content of 1.5 % and B-unit content of 15.0 %

\*\* Tetrakis [methylene (3,5-di-*tert*-butyl-4-hydroxy-hydro-cinnamate) methane

Table 3. Matte-Translucent Film Layer Compositions of Present Invention

Exp. No.	C o m p o s i t i o n   o f   L a y e r s				
	A	B	C	D	E
E 1	3 $\mu$ m [E(3.5%)-P-B(5.5%) + PP + HDPE + SS + EA + APH + Cast]	0.5 $\mu$ m [PP + APH + TAM + Cast]	13 $\mu$ m [PP + APH + TAM + Cast]	0.5 $\mu$ m [PP + APH + TAM + Cast]	0.5 $\mu$ m [PP + APH + SS + Cast]
E 2	3 $\mu$ m [E(3.5%)-P-B(5.5%) + PP + HDPE + SS + EA + APH + Cast]	0.5 $\mu$ m [PP + APH + TAM + Cast]	45.5 $\mu$ m [PP + APH + TAM + Cast]	0.5 $\mu$ m [PP + APH + TAM + Cast]	0.5 $\mu$ m [E(4.2%)-P-B(9%) + PP + HDPE + SS + EA + APH + Cast]
E 3	3 $\mu$ m [E(3.5%)-P-B(5.5%) + PP + HDPE + SS + EA + APH + Cast]	0.5 $\mu$ m [PP + APH + TAM + Cast]	25.5 $\mu$ m [PP + APH + + TAM + Cast + EtA]	0.5 $\mu$ m [PP + APH + TAM + Cast]	0.5 $\mu$ m [PP + APH + SS + Cast]
E 4	3 $\mu$ m [E(3.5%)-P-B(5.5%) + PP + HDPE + SS + EA + APH + Cast]	0.5 $\mu$ m [PP + APH + TAM + Cast] SS + Cast]	25.5 $\mu$ m [PP + APH + EtA + TAM + Cast]	0.5 $\mu$ m [PP + APH TAM + Cast]	0.5 $\mu$ m [E(1.5%)-P-B(15%) + APH + EA + + SS + Cast]
E 5	3 $\mu$ m [E(3.5%)-P-B(5.5%) + PP + HDPE + SS + EA + APH + Cast]	-	26 $\mu$ m [PP + APH + TAM + Cast]	0.5 $\mu$ m [PP + APH + TAM + Cast]	0.5 $\mu$ m [PP + APH + SS + Cast]
E 6	3 $\mu$ m [E(3.5%)-P-B(5.5%) + PP + HDPE + SS + EA + APH + Cast]	-	26 $\mu$ m [PP + APH + TAM + Cast + EA + EtA]	0.5 $\mu$ m [PP + APH + TAM + Cast]	0.5 $\mu$ m [PP + APH + SS + Cast]
E 7	3 $\mu$ m [[E(3.5%)-P-B(5.5%) + PP + HDPE + SS + EA + APH + Cast]	-	26 $\mu$ m [PP + APH + TAM + Cast]	0.5 $\mu$ m [PP + APH TAM + Cast]	0.5 $\mu$ m [E(1.5%)-P-B(15%) + APH + EA + + SS + Cast]
E 8	3 $\mu$ m [E(3.5%)-P-B(5.5%) + PP + HDPE + SS + EA + APH + Cast]	-	13 $\mu$ m [PP + APH + TAM + Cast]	-	0.5 $\mu$ m [PP + APH + SS + Cast]
E 9	3 $\mu$ m [E(4.2%)-P-B(9%) + PP + HDPE + St-MA SS + EA + APH + Cast]	0.5 $\mu$ m [PP + APH + TAM + Cast]	13 $\mu$ m [PP + APH + TAM + Cast]	0.5 $\mu$ m [PP + APH + TAM + Cast]	0.5 $\mu$ m [PP + APH + SS + Cast]
E 10	3 $\mu$ m [E(4.2%)-P-B(9%) + PP + HDPE + PMMA SS + EA + APH + Cast]	0.5 $\mu$ m [PP + APH + TAM + Cast]	13 $\mu$ m [PP + APH + TAM + Cast]	0.5 $\mu$ m [PP + APH + TAM + Cast]	0.5 $\mu$ m [PP + APH + SS + Cast]
E 11	3 $\mu$ m [E(3.5%)-P-B(5.5%) + PP + HDPE + DIB-MA + EA + APH + Cast]	1.5 $\mu$ m [PP + APH + TAM + Cast]	43 $\mu$ m [PP + APH + TAM + Cast]	1.0 $\mu$ m [PP + APH + TAM + Cast]	1.5 $\mu$ m [PP + APH + SS + Cast]

**Table 4. Properties of Biaxially Oriented Polypropylene Based  
Matte-Translucent Films of Present Invention**

Properties E11	Example Numbers										
	E1	E2	E3	E4	E5	E6	E7	E8	E9	E10	
Thickness ( $\mu\text{m}$ )	17.5	50	30	30	30	30	30	15	17.5	17.5	50
Amount of layers	5	5	5	5	4	4	4	3	5	5	5
Density ( $\text{g/cm}^3$ )	0.92	0.90	0.90	0.90	0.90	0.91	0.90	0.89	0.92	0.92	0.91
Sheen ( angle $85^\circ$ )											
Skin matte surface	13	15	15	15	14	13	15	13	10	11	10
Skin shining surface	43	-	61	60	44	42	56	43	60	58	60
Haze ( % )	55	60	70	70	57	55	62	55	65	70	70
Coefficient of friction	0.5	0.4	0.3	0.5	0.5	0.5	0.6	0.5	0.5	0.4	0.3
Shrinkage ( % )											
90°C in LS	0.5	0.8	1.25	1.2	0.6	0.5	0.5	1.0	0.4	0.5	0.4
90°C in TS	0.25	0.3	0.5	0.5	0.2	0.25	0.4	0.25	0.25	0.2	0.2
120°C in LS	2.5	3.5	5.5	5.6	2.5	2.2	2.5	3.0	2.5	3.0	3.0
120°C in TS	1.5	2.0	2.25	2.22	1.5	1.3	2.0	1.6	1.5	2.0	1.8
Tensile strength ( $\text{kg/mm}^2$ ) in LS	22	20	15	18	20	21	12	10	25	23	24
in TS	25	30	30	29	25	26	25	21	61	59	60
Elongation ( % )											
in LS	135	145	170	165	140	138	150	135	135	135	135
in TS	55	45	61	61	60	50	52	45	43	45	43
Minimum heat sealing temperature ( $^\circ\text{C}$ )	110	105	105	70	100	105	110	95	105	105	110
Matte appearance (visual assessment)	+++	+++	++	+++	+++	++	+++	+++	++	—	++
Surface tension* ( $\text{mN/m}$ )	40	41	41	40	40	41	41	40	41	41	40

\* Measured after storage of corona discharged films for 6 months.

## Claims

1. A five-layer biaxially oriented polypropylene based matte-translucent film with thickness of 15-50  $\mu\text{m}$  comprise (A) 1.5-3.0  $\mu\text{m}$  skin matte-translucent layer containing ethylene(E)-propylene(P)-butylene(B) terpolymer with E-unit content of 1.5-4.9 % and B-unit content of 5.5-15 %, polypropylene, high density polyethylene (HDPE) and/or 10: (0.5-1.5) mixture or blend of HDPE with polymethyl(metha)acrylates, alternating or random copolymer of vinyl monomer (styrene, alkyl(metha)acrylates, etc.) or  $\alpha$ -olefins ( $\text{C}_{2-10}$ ) with anhydrides or esters of unsaturated mono- or dicarbonic acids as mattering agent, synthetic silica as an antiblocking agent, erucicamide as a slip agent and allyl phosphite (APH) as antioxidants, calcium stearate as a neutralizer, and tetrakis [methylene (3,5-di-*tert*-butyl-4-hydroxy-hydro-cinnamate)] methane as a stabilizer, (B)  $\geq 0.5 \mu\text{m}$  polypropylene inner layer containing said APH antioxidant, stabilizer, neutralizer and/or slip agent, and/or  $\geq 1.5 \mu\text{m}$  inner layer with composition as a (A) matte layer, (C)  $\leq 45.5 \mu\text{m}$  virgin or marked polypropylene core layer containing 5-cholesten-3 $\beta$ -ol as a marked agent, saids APH antioxidant, stabilizer and neutralizer, and/or said slip agent and ethoxylated amine as a antistatic agent, (D)  $\geq 1.5 \mu\text{m}$  polypropylene inner layer containing said APH antioxidant, stabilizer and neutralizer, and (E)  $\geq 0.5 \mu\text{m}$  outer shining layer containing polypropylene, said antiblocking agent, APH antioxidant and neutralizer, and/or  $\geq 1.5 \mu\text{m}$  outer matte layer with composition as a (A) layer, were prepared by using the tandem extruder system supplied with three or four satellite co-extruders, flat die, chill roll, and recycling line. Biaxially oriented film (longitudinal stretching at two steps by overall factor of 6-7 times and transverse stretching of 9-10 times) has a sheen of less than or equal to about 15 (angle 85°), a longitudinal shrinkage (15 min) of less than or equal to about 1.2 % at 90°C and about 5.5 % at 120°C, and a transverse shrinkage (15 min) of less than or equal to about 0.5 % at 90°C and about 2.2 % at 120°C, heat-sealing temperature of or equal to about 70-110°C,

excellent matte appearance ( + + + visual assessment) and surface tension of equal to about 40-41 *mN/m* (after storage for 6 months).

2. A film according to claim 1 is contained 10:(0.5-1.5) mixture and/or blend of  
5 HDPE / alternating styrene-maleic anhydride (St-MA, Exp.9) or  $\alpha$ -diisobutylene-maleic anhydride (DIB-MA, Exp.10) copolymers, or HDPE / polymethylmethacrylate (PMMA, Exp.11), and other carbonyl containing alternating or random copolymer and/or homopolymer.
3. A film according to claim 1 contains tetrakis [methylene (3,5-di-*tert*-butyl-4-  
10 hydroxy-hydro-cinnamate)] methane as a stabilizer and allyl phosphite as an antioxidant.
4. A film according to claim 1, wherein film possessed excellent matte appearance provided with given composition and structure of its.
- 15 5. A matte-translucent skin layer of the film according to claim 1 have lower value of sheen of less than or equal to about 15 (angle 85°).
6. A film according to claim 1 have low value of longitudinal shrinkage (15 *min*)  
20 of less than or equal to about 1.2 % at 90°C and about 5.5 % at 120°C, and low value of transverse shrinkage (15 *min*) of less than or equal to about 0.5 % at 90°C and about 2.2 % at 120°C.
7. A film according to claim 1 have high surface tension value of equal to about  
25 40-41 *mN/m* after storage for 6 months.
8. A film according to claim 1 have a minimum heat-sealing temperature of equal to about 70-110°C.



9. A film according to claim 1 is prepared by using the tandem extruder system supplied with three or four satellite co-extruders, recycling line, flat die and air corona discharge including following stages of production process: (1) extrusion is carried out at 200-280°C and taking the co-extruded film off over at first chill  
5 roll temperature of 20-40°C, (2) longitudinal stretching ratio is from 1.5:1 to 7:1 at 100-160°C, (3) transverse stretching ratio is from 7:1 to 10:1 at 160-185°C, (4) setting process is produced at 165-170°C, and (5) air corona discharge of heat film is carried out at voltage of 15-25 kV and frequency of 20-30 kHz.
- 10 10. A film according to claim 9 is prepared by using modified process of chill-roll treatments in two steps of the biaxially orientation allowing to form good homogenized matte film with improved surface properties and dimensional stability.
- 15 11. A process of production of film according to claim 9 is included a recycling process of film waste forming in the transverse stretching stage. It is claimed that use of said recycling is decreased film cost by 3.5 %.
- 20 12. The films according to claims 1-11 are recommended for photographic paper laminations, reproduction pictures and menu coatings, conventional lamination purposes, hygienic applications, health care products, food stuffs packaging, cosmetics and frames, high speed packaging machines, and as matte tape base films, and for general packaging and overwrapping applications.

# INTERNATIONAL SEARCH REPORT

International application No.  
PCT/KR 98/00017

## A. CLASSIFICATION OF SUBJECT MATTER

IPC<sup>6</sup>: B 32 B 27/32, 27/30; B 65 D 65/40; B 29 D 9/00

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC<sup>6</sup>: B 29 D; B 32 B; B 65 D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EPODOC, PAJ, WPI

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 619 183 A1 (HOECHST AKTIENGESELLSCHAFT) 12 October 1994 (12.10.94), claims; page 10, lines 5-31	1,2,4,9,12
A	EP 0 616 885 A1 (WOLFF WALSRODE AKTIENGESELLSCHAFT) 28 September 1994 (28.09.94), claims.	1,2,4,9,10,12
A	EP 0 538 746 A1 (HOECHST AKTIENGESELLSCHAFT) 28 April 1993 (28.04.93), claims; page 4, lines 6-33.	1,2,4,9,10,12
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☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

### \* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

13 October 1998 (13.10.98)

Date of mailing of the international search report

28 October 1998 (28.10.98)

Name and mailing address of the ISA/

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Kohlmarkt 8-10; A-1014 Vienna  
Facsimile No. 1/53424/535

Authorized officer

Weigerstorfer

Telephone No. 1/53424/221

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/TR 98/00017

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